



# **RADIOISOTOPES (RA-226, TH-232, K-40) CONCENTRATION ASSESSMENTS IN THE IMPORTED MARBLES USED FOR BUILDING PURPOSES IN NIGERIA**

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## **ABSTRACT**

*Radionuclides (Ra-226, Th-232, K-40) concentrations were evaluated for six brands of imported marbles used in Nigeria using Hyper Purity Germanium Gamma detector (HPGe detector). The results showed that the concentration of radioisotope presents ranged between 4.30 and 94.47 Bqkg<sup>-1</sup>; 42.09 and 104.28 Bqkg<sup>-1</sup>; and 181.40 and 871.72 Bqkg<sup>-1</sup> for <sup>226</sup>Ra, <sup>232</sup>Th and <sup>40</sup>K, respectively. The results revealed that the radium equivalent measures in the samples were found to be within the recommended level. The value of dose rates obtained ranged between 34.94 and 135.68 nGyh<sup>-1</sup>. Highest values of annual effective dose rate of about 1.420, 1.396 and 1.138 mSv y<sup>-1</sup> were observed in Rose Bite, Black Galaxy and Blue Pearl marbles respectively. The obtained value for internal ( $H_{in}$ ) and external hazard ( $H_{ex}$ ) indices ranges from 0.223 to 1.077 and 0.212 to 0.821 respectively. The gamma ( $I_\gamma$ ) and alpha ( $I_\alpha$ ) activities indices estimated ranged between 0.285 and 1.098 and 0.022 and 0.472 respectively. The activity utilization index is estimated to be 5.547 and is noted in Rose Bite marble while the lowest value of 1.289 is observed in Green Pearl marble. The estimated life*

*time cancer risk for the measured marble samples is above the permissible limit value of 0.00029. As a result of this, marbles used in this study may mainly be useful for exterior decoration purposes when needed for building project in order to avoid health risk associated with it.*

**Key words:** Radioisotopes; HPGe; Imported-marbles; Hazard-indices.

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## 1. INTRODUCTION

Building materials often have traces of naturally occurring radioactivity, which mostly come from radionuclides such as  $^{238}\text{U}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  series[1]. But among the series of uranium chain, the most useful in terms of radiological assessment is  $^{226}\text{Ra}$  [2] which is always used as reference instead of uranium. The radioisotopes in the building materials may result to both internal and external emissions which may affect the occupants of the buildings where these materials are used. The internal exposure is mainly caused by the radioactive inert gas called radon while external exposure is as a result of direct gamma ray radiation originating from the building constructing materials [3]. Globally, the mean concentrations of radioactive materials in the earth's crust for radionuclides such as radium, thorium and potassium are 35, 30 and 400  $\text{Bqkg}^{-1}$  respectively [2]. The global average of indoor effective dose as a result of gamma emission emanating from building constructing materials has been approximately valued to be within the range of  $0.4 \text{ mSvy}^{-1}$ . But 80 percent of human beings spend more time indoors and are thus exposed in long-term to the radiation that occurs internally and externally through building materials such as ceramic tiles, gravel, marbles etc[4-5]. Therefore, this significant amount of gamma dose rate emanating from building materials may be as a result of natural radioactive contents in the radionuclides materials[4]. Various international organizations and groups of individual have undertaken research relating to ionizing radiation associated with building materials with the purpose of assessing its radiological impacts [6, 7, 8, 9, 10, 11, 12, 13, and 14]. Various natural construction materials used extensively for building purposes include sediment from river (sand), coal slag, gypsum etc for plastering, bricks mending and flooring alongside the new building products which may act as substitutes during building processes but the new products of building materials may consist of high level natural radioactivity. However, knowledge of these levels of natural radioactivity in building materials and its probable radiological hazards indices is necessary in order to ensure human safety in terms of health and to establish measures and suggestions in the management and use of these materials. Geologically, marble is a kind of metamorphic rock that consists of crystalline quantity of dolomite. As a result of its smooth surface and existence in various attractive colours, it is broadly used as building materials. Mostly, marble is an extraction from the mountains in the source countries[15]. This study is aimed at investigating the radioactivity concentration levels in most used marbles imported in Nigeria for building purposes.

## 2. MATERIALS AND METHODS

### 2.1. Preparation of Samples

Six samples of various marbles (Table 1) namely: Black Galaxy (BG), Blue Pearl (BP), Green Pearl (GP), Jubrano (Ju), Rose Bite (RB) and Tam Bite (TB) marbles of different brands which were imported to Nigeria were bought from different suppliers and prepared according to IAEA standard [16]. These marbles were purchased from Nigerian commercial market (OrileIganmu in Lagos State, Nigeria). Labeling was done for the purpose of easy recognitions. The marbles were smashed into small pieces for further action. All the sample materials used were shattered through the use of laboratory milling engineering machine. Thereafter the laboratory machine was intensively cleansed with the use of high pressure blower. This is done in order to eliminate the contamination of each sample used. The whole processes were recapitulated for each sample till all the samples were thoroughly grinded into powdered form. The disk grinder made by Christy & Norris Limited was used for the pulverizing. The crushed samples were made to pass through 250  $\mu\text{m}$  sieve mesh size; afterward 1 kg of the sample sieved was measured and put in high density polyethylene bottle, well labeled with indelible marker. Each of the samples was measured again by using digital balance of detection limit of  $\pm 0.01$  g differently into the Marinelli beakers. The samples were sealed in Marinelli beakers for 4 weeks secular equilibrium for the impression that the parent nuclide is equal to the daughter. The Marinelli beaker used in this analysis is for both samples and IAEA standard of which they are of the same geometry in terms the detector size.

**Table 1.** The list for some of imported marbles from India

Brands Name	Brands Name Code	Sample Size (mm)	Country
Black Galaxy	BG	(600X 300)mm	India
Blue Pearl	BP	(600X 300mm)	India
Green Pearl	GP	(600X 300mm)	India
Jubrano	JU	(600X 300mm)	India
Rose Bite	RB	(600X 300mm)	India
Tam Brown	TB	(600X 300mm)	India

### 2.2. Gamma Spectrometric Analysis of the Selected Samples

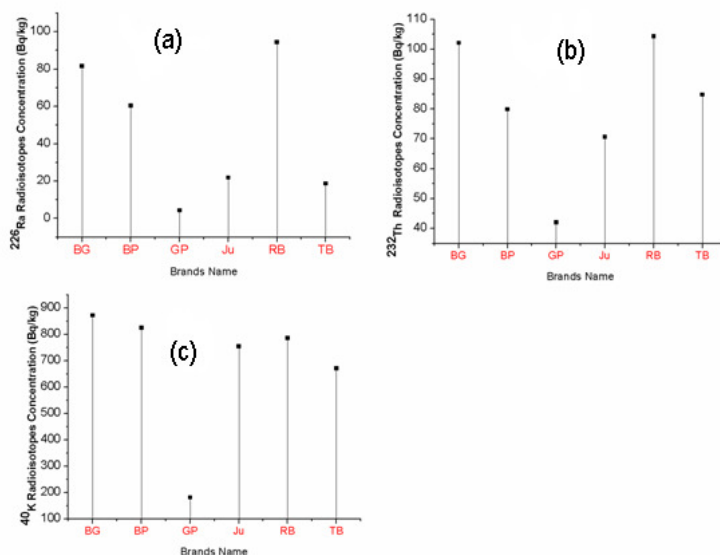
Analysis of the samples were conducted in Canada Activation Laboratory System using High Purity Germanium detector, Canberra Lynx<sup>TM</sup> Digital Signal Analyzer (DSA), a 32 K channel integrated signal analyzer and a top-opening lead shield (4" lead, copper/tin liner) to prevent high background counts with 50 % relative efficiency and resolution of 2.1 keV at 1.33 MeV gamma energy of <sup>60</sup>Co. The software used in locating and analyzing the peaks is called Genie-2K V3.2 and this was used for the subtraction of background counts and identification of the nuclides. The proficiency curves for this investigation were rectified for both self-absorption and attenuation effects of the emission of gamma photons. CAMET and IAEA standards (DL-1a, UTS-2, UTS-4, IAEA-372 and IAEA-447) were used for checking the efficiency and effectiveness of the calibration system. For the activity measurements, the samples were counted for 86,400 seconds with the background counts subtracted from the net count. The minimum level of detectable activity of the detector was fixed with a confidence level of 95 %. The uncertainties in errors value were estimated keeping into account the related errors connected to the gamma efficiency and probability calibration standard of the system. The emitting gamma line of 609 keV, 934 keV, 2204 keV, 1764 keV and 351 keV, 295 keV were

used for the progeniture of radium ( $^{214}\text{Bi}$  and  $^{214}\text{Pb}$ ) but 1764 keV was the emission resolution used for radium due to its low self-attenuation effects at high energy. In as much as  $^{232}\text{Th}$  cannot be detected directly, the activity of its progeniture ( $^{208}\text{Tl}$  and  $^{228}\text{Ac}$ ) with the use of 2614.53 keV, (35.63%), 583 keV (30.3%) and 911 keV, 338 keV, 463 keV was estimated while 1461 keV (10.7%) was used in resolving  $^{40}\text{K}$  [5].

### 3. RESULT AND DISCUSSIONS

#### 3.1. Radioisotope (Ra-226, Th-232, K-40) concentration measurement

The radionuclides concentrations estimated in the imported marbles are shown in Figure 1a – 1c. The activities concentration of the radioisotopes present in the marbles ranged between 4.30 to 94.47 Bqkg<sup>-1</sup> with highest value of 94.47 Bqkg<sup>-1</sup> noticed in Rose Bite (RB) marble and lowest value observed in Green Pearl (GP) marble for  $^{226}\text{Ra}$ . For  $^{232}\text{Th}$ , the radioisotopes concentrations varies from 42.09 to 104.28 Bqkg<sup>-1</sup>, the lowest value of 42.09 Bqkg<sup>-1</sup> is observed in Green Pearl (GP) marblesample while the highest is noted in the Rose Bite (RB) with value of 104.28 Bqkg<sup>-1</sup> and followed by Black Galaxy (BG) marble. Furthermore,  $^{40}\text{K}$  concentrations (Figure 1c) in the marbles ranged between 181.40 and 871.72 Bqkg<sup>-1</sup> with highest value noted in Black Galaxy (BG) while lowest value of 181.40 Bqkg<sup>-1</sup> observed in Green Pearl (GP). The estimated mean values for  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  are 46.88, 80.64 and 681.39 Bqkg<sup>-1</sup> respectively and these obtained values are quite high compared to the international recommended values of 35, 30, and 400 Bqkg<sup>-1</sup> [6]. This higher values of  $^{40}\text{K}$  observed in the samples especially Black Galaxy (BG), Blue Pearl (BP), Jubrano (Ju), Rose Bite (RB) and Tam Brown (TB) (Figure 1c), could be as a result of ion-exchange of clay contents together with a simultaneous sedimentation of mineral particles containing uranium and thorium which could be associated to the higher activity concentrations determined [16-17]. It was observed that there is consistency in the lowest value of radioactivity concentration ( $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$ ) obtained for Green Pearl (GP) marble while there are variations in other samples (Figure 1a – 1c).



**Figure 1.** Variation in radioisotopes concentration of (a)  $^{226}\text{Ra}$ , (b)  $^{232}\text{Th}$  and (c)  $^{40}\text{K}$

### 3.2. Radiological risk assessment

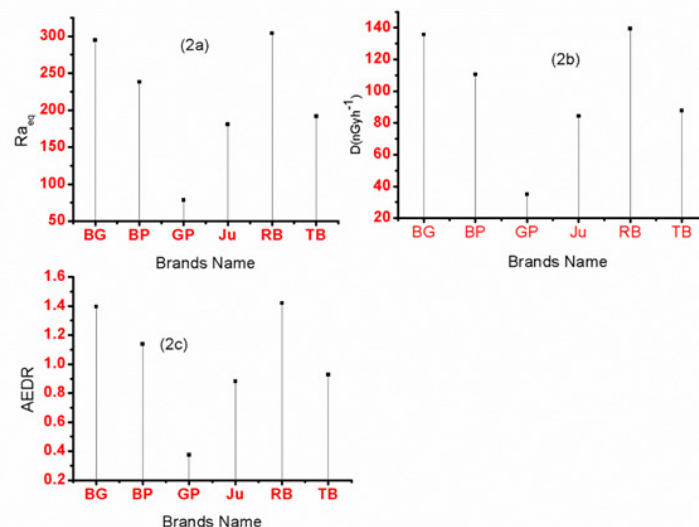
#### 3.2.1. Estimation of radium equivalent ( $Ra_{eq}$ ) activity

In this present investigation, the estimation of radium equivalent ( $Ra_{eq}$ ) activity is needed in the samples analysed in order to compare the radioactivity contents present in the samples which consists of  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$ . Radium equivalent ( $Ra_{eq}$ ) was used as a common factor to establish the sum of the concentrations of the activities.  $Ra_{eq}$  activities was estimated based on the international reference value of  $370 \text{ Bqkg}^{-1}$  for  $^{226}\text{Ra}$ ,  $259 \text{ Bqkg}^{-1}$  for  $^{232}\text{Th}$  and  $4810 \text{ Bqkg}^{-1}$  for  $^{40}\text{K}$  [6]. Radium equivalent with unit in  $\text{Bq/kg}$  was calculated using equation (1).

$$R_{aeq} = C_{Ra} + 1.43C_{Th} + 0.077C_K \quad (1)$$

where  $C_{Ra}$ ,  $C_{Th}$  and  $C_K$  are the average activity concentrations of radio nuclides determined in the samples used in  $\text{Bq kg}^{-1}$  respectively.

In consideration of Figure 2a, it was observed that the highest value of radium equivalent of  $304.09 \text{ Bqkg}^{-1}$  was found to be Rose Bite marble and the lowest value of  $78.46 \text{ Bqkg}^{-1}$  is noted in Green Pearl marble. The mean value of radium equivalent which is  $214.65 \text{ Bqkg}^{-1}$  is within the international reference value of  $370 \text{ Bqkg}^{-1}$ . Generally, the radium equivalent for each of the marbles is within the recommended limit value (Figure 2a).



**Figure 2.** Radiological risk parameters: (a)  $Ra_{eq}$ , (b)  $D (\text{nGyh}^{-1})$  and (c)  $AEDR (\text{mSvy}^{-1})$

#### 3.2.2. The Absorbed Dose Rate ( $Dr$ )

In this present investigation, the dose rates absorbed ( $Dr$ ) were determined from the calculated radioactivity concentrations as shown in Figure 2b. The sum of air dose rates ( $Dr$ ) present in an open air above 1 m on the ground is as a result of gamma ray emission which emits from the radionuclides of  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  in  $\text{BqKg}^{-1}$ . Equation (2)[6] was used to determine the Dose rate

$$D_r = 0.462C_{Ra} + 0.604C_{Th} + 0.0417C_K \quad (2)$$

where  $C_{Ra}$ ,  $C_{Th}$  and  $C_K$  are the average activity concentrations of evaluated radionuclides in  $\text{Bq kg}^{-1}$  respectively.

The value of dose rates ranged between  $34.94$  and  $156.40 \text{ nGyh}^{-1}$ . The highest value of dose rate was noticed in Rose bite marble with value of  $135.68 \text{ nGyh}^{-1}$  while the lowest value was found to be Green Pearl marble with value of  $34.94 \text{ nGyh}^{-1}$ . In comparison of absorbed

dose rate in this present study with international reference limit, it was observed that the calculated value for all the marbles sample used were higher than the international reference value of  $80 \text{ nGy h}^{-1}$  as reported by [6] except the Green marble which has value of  $34.94 \text{ nGy h}^{-1}$ . This is shown in Figure 2b.

### 3.2.3. The Annual Effective Dose Rate (AEDR)

The indoors effective dose rate which is usually experienced by human beings from the internal dose rate in terms of occupancy factor is elucidated as the human occupancy level in an area closest to source of radiation is established as 80 percent of 8760 hours per year, and the transformation factor of  $0.7 \text{ Sv Gy}^{-1}$  is usually used in the conversion of the absorbed dose rate in the air to effective dose [6]. The yearly effective dose is evaluated using the application of equation (3).

$$AEDR = (0.49C_{Ra} + 0.76C_{Th} + 0.048C_K) \times 8.76 \times 10^{-3} \quad (3)$$

where  $C_{Ra}$ ,  $C_{Th}$  and  $C_K$  are the average activity concentrations of evaluated radionuclides in  $\text{Bq kg}^{-1}$  respectively.

The annual effective dose rate for the imported marbles ranged between  $0.375$  and  $1.420 \text{ mSv y}^{-1}$ . The high values of  $1.420$ ,  $1.396$  and  $1.138 \text{ mSv y}^{-1}$  is observed in Rose bite, Black Galaxy and Blue pearl marbles respectively while the lowest value of  $0.375 \text{ mSv y}^{-1}$  is found in Green pearl marble as shown in Figure 2c. In comparing the yearly effective dose rate of brands sample of marbles with international reference value of  $0.07 \text{ mSv y}^{-1}$ , it was observed that most of the brands of the marbles used have very high value of AEDR except the Green pearl marble.

### 3.2.4. Determination of Internal Hazard Index ( $H_{in}$ )

The internal hazard ( $H_{in}$ ) which is defined in relationship to hazard indices can be estimated by the use of equation (4) [18]:

$$H_{in} = \frac{C_{Ra}}{185} + \frac{C_{Th}}{259} + \frac{C_K}{4810} \quad (4)$$

Where  $eC_{Ra}$ ,  $C_{Th}$  and  $C_K$  are concentrations of natural radioactivity of  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  in  $\text{Bq kg}^{-1}$ . In order to ascertain the safety of using building materials such as marble for construction purposes, the internal hazard index value should be less than 1. From Figure 3, the value ranges from  $0.223$  to  $1.077$  with Rose Bite and Black Galaxy having the highest value of  $1.077$  and  $1.016$  which is higher than international reference value of unity. The other brands of marbles have value that is less than unity but the lowest value of  $H_{in}$  is found in Green Pearl marble. It was observed further that Rose Bite and Black Galaxy marbles may not be the best choice for indoor or internal usage in terms of decoration or construction purposes.

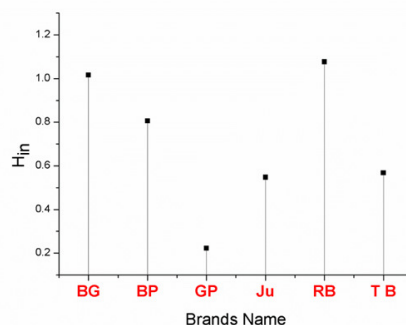


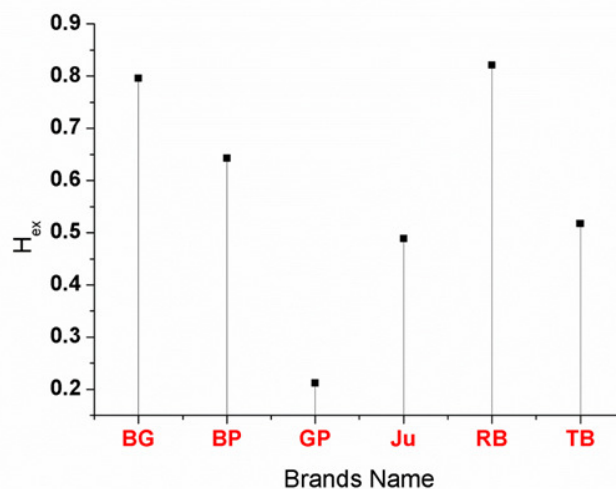
Figure 3. Variation in internal hazard index in samples used

### 3.2.5. Evaluation of External Hazard Index ( $H_{ex}$ )

The emission of gamma radiation hazard index due to the prescribed natural radioactivity was appraised by external hazard radiation and evaluated by applying equation (5)[6].

$$H_{ex} = \frac{C_{Ra}}{370} + \frac{C_{Th}}{259} + \frac{C_K}{4810} \quad (5)$$

Where  $C_{Ra}$ ,  $C_{Th}$  and  $C_K$  are the concentrations of activities of  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  in  $\text{Bqkg}^{-1}$  respectively. The values of  $H_{ex}$  ranged between 0.212 and 0.821 with the highest value of 0.821 noted in Rose Bite marble whereas the lowest value of 0.212 is noticed in Green Pearl marble. Generally, it was observed that the values of  $H_{ex}$  in all the samples were less than unity as recommended by [6] suggesting that marbles could be used for outside construction purposes in terms of decoration which may not pose any health threat. This is shown in Figure 4.



**Figure 4.** Variation in external hazard index in samples used

### 3.2.4. Gamma Index Determination ( $I_\gamma$ )

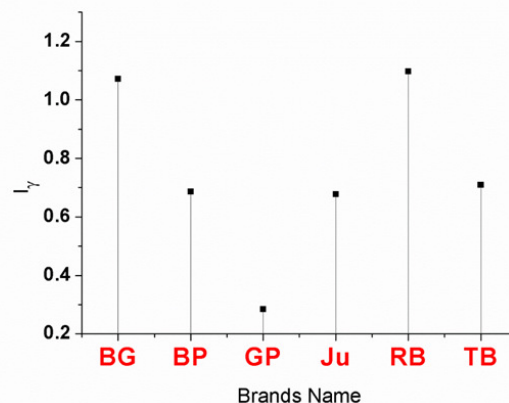
Gamma ray index is another important radiological hazard that can be used to estimate the  $\gamma$ -emission risk relating to the natural radioactive materials in the specific varieties under study. The gamma ray index representative ( $I_\gamma$ ) is calculated using equation (6) as reported by [19].

$$I_\gamma = \frac{C_{Ra}}{300} + \frac{C_{Th}}{200} + \frac{C_K}{3000} \quad (6)$$

Where  $C_{Ra}$ ,  $C_{Th}$  and  $C_K$  are concentrations of natural radioactivity of  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  in  $\text{Bqkg}^{-1}$ . The check on the radio nuclides of building construction or decorating materials as reported by Righi and Bruzzi[20] is dependent on standard level of dose used for exemption and control. The effective doses that are greater than the standard level of  $1 \text{ mSvy}^{-1}$  should be considered in terms of protection of emission. Recommendation of dose that ranged between  $0.3$  and  $1 \text{ mSvy}^{-1}$  was considered as a control, which serves as the excessive gamma dose to that absorbed outdoors. The gamma emission activities index is useful in the identification of whether a criterion dose is met [20]. This gamma emission activities index describe the processes and aggregate in which the building materials is used, with the limitation of their value indices not greater than the reference value and is dependent on the criterion dose level as displayed in Figure 5. In this present study the dose evaluated has excluded the doses at the background level that has been protected by the building materials that was used in bulk but this do not exempt building materials when used as an exterior material because the layers of



the exterior materials that is thin does not lower appreciably the dose at the background level. The gamma emission activities index  $\leq 1$ , proportional to the annual effective dose lower than or equate to  $1 \text{ mSvy}^{-1}$ , while gamma emission activity index  $\leq 0.5$  equals  $0.3 \text{ mSvy}^{-1}$  if the quantity of the materials are used in bulk. Moreover, gamma emission activities index  $\leq 6$  equate to annual effective dose of  $1 \text{ mSvy}^{-1}$  and gamma emission activities index  $\leq 2$  equivalent to an annual effective dose  $\leq 0.3 \text{ mSvy}^{-1}$  when the vastness of the materials are applied in an exterior manner [20]. The results of gamma emission activities index is shown in Figure 5. The values ranges from 0.285 to 1.098 with highest value of 1.098 reported in Rose Bite marble followed by BlackGalaxymarbles with value of 1.072 while the lowest value of 0.285 is found in Green Pearl marble. In comparing the estimated value of gamma emission index with international reference value when the building materials is applied externally, it was observed thatIfy value for Black Galaxyand Rose Bite are within the range whereas in terms of using the material in bulk form marbles such as Blue Pearl, Green Pearl, Jubranoand Tam Brown are within the range of international reference limit.



**Figure 5.** Variation on the Gamma activity index obtained in the marbles used

### 3.2.5. Determination of Alpha Index (I<sub>α</sub>)

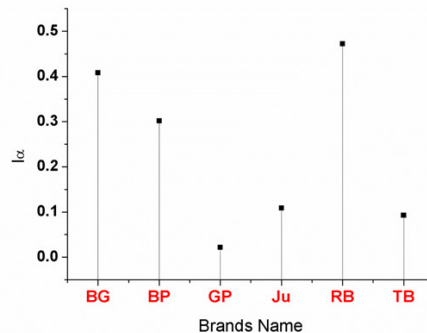
The evaluation of alpha index is one of the cogent aspects of radiological hazard riskanalysis that is concerned with the estimate of excess of alpha emission radiation as a result of radon inhalation which originates from materials used for constructing buildings such as marble. The alpha index is calculated using equation (7) [21] is:

$$I_{\alpha} = \frac{C_{Ra}}{200}(7)$$

Where  $C_{Ra}$  is the radium concentration in  $\text{Bqkg}^{-1}$  for building materials. If the activity of radium level in building material is far beyond  $200 \text{ Bqkg}^{-1}$  there is probability that the radon-exhalation emanating through the material maypose concentration of indoor radon. Figure 6 displays the variations in the activity of alpha index. The ICRP suggested an activity level of  $200 \text{ Bqm}^{-3}$  as a standard value radon for dwelling house [22]. Furthermore, if the activity level of radium is less than  $100 \text{ Bqkg}^{-1}$ , it reveals that radon-exhalation emanating through the building materials may likely not cause indoor concentration that is above  $200 \text{ Bqm}^{-3}$ [21]. It has been documented that the suggested excluded value and the suggested upper limit for concentrations of radon are  $100 \text{ Bqkg}^{-1}$  and  $200 \text{ Bqkg}^{-1}$  when dealing with building materials [23] and the upper limit for concentrations of radon (I<sub>α</sub>) is equivalent to 1 [24]. This is shown in Figure 6. In this study, the results showed that concentrations of radon in marbles ranged between 0.022 and 0.472 with high values of radon concentration noted in Rose Bite and



Black Galaxy marbles respectively while lowest is observed in Green Pearl marble followed by Tam Brown and Jubrano marbles.



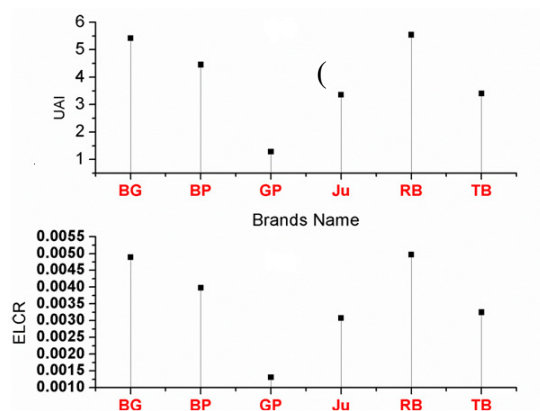
**Figure 6.** Variation on the Alpha activity index obtained in the marbles used

### 3.2.6. Utilization Activity Index (UAI)

The use of the utilization activity index (UAI) for building materials can be estimated through the sum of the radionuclides such as  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  and this can be estimated by the use of equation (8) [25]:

$$AUI = \left( \frac{C_{Ra}}{50} \right) f_{Ra} + \left( \frac{C_{Th}}{50} \right) f_{Th} + \left( \frac{C_K}{500} \right) f_K \quad (8)$$

Where  $C_{Th}$ ,  $C_{Ra}$ , and  $C_K$  are the true figures of the activity in a unit of mass ( $\text{Bqkg}^{-1}$ ) of  $^{232}\text{Th}$ ,  $^{226}\text{Ra}$ , and  $^{40}\text{K}$  independently as analyzed in the building constructing materials such as marble.  $f_{Ra}$ ,  $f_{Th}$  and  $f_K$  are the contributions of the sum of dose rate at fractional level and is associated to gamma emission from the concentration of the actual activities of the studied radioactive materials. In order to ascertain the contribution of utilization activity index in building material, the estimated value should be less than 2 and is equivalent to a dose rate of  $80 \text{ nGyh}^{-1}$  [26]. The utilization index values for the marbles used in this present study ranges from 1.289 to 5.547. The lowest value of 1.289 is observed in Green Pearl marble while the highest value of 5.547 is noted in Rose Bite. Other marbles that have utilization index value that is higher than international reference value of 2 are Black Galaxy, Blue Pearl, Tam Brown and Jubrano in that decreasing order and Figure 7a shows the variation in the utilization activity index values.



**Figure 7.a)** Utilization Activity Index and b) Excess Lifetime Cancer Risk

### 3.2.7. Excess Lifetime Cancer Risk (ELCR)

The lifetime cancer risk is of one of the hazard parameters evaluated in this present study in order to ascertain the kinds of health implications associated with the use of marbles for building construction purposes and was determined using equation 9 [26,27]. The ELCR is represented as:

$$ELCR = AEDR \times DL \times RF \quad (9)$$

where AEDR is annual effective dose rate, DL is duration of life (70 years) and RF is risk factor ( $0.05 \text{ Sv}^{-1}$ ).

The kind of risk factor used in this study is the fatal cancer risk per Sievert which is generally used for the public with the value of  $0.05 \text{ Sv}^{-1}$  [26, 28]. The estimated values of ELCR observed in this investigation ranged between 0.00131 and 0.00497. The highest value of 0.00497 was observed in Rose Bite marble followed by Black Galaxy marble but the least value of 0.00131 was noticed in Green Pearl marble as shown in Figure 7b. Comparing each of the marbles used with the international reference value, it was observed that all the samples have ELCR value that is higher than the mean reference limit value of 0.00029 [6].

### 3.2.8. Comparison between Utilization Activity Index (UAI) and Excess Life Cancer Risk (ELCR)

In comparing the UAI and ELCR, it was observed that Rose Bite (RB) marble has utility activity index of value 5.547 and ELCR value of 0.00497 whereas the lowest values of 1.289 and 0.00131 for both UAI and ELCR were recorded in Green Pearl (GP) marble. The other higher values for UAI and ELCR were observed in marbles such as Black Galaxy (BG), Blue Pearl (BP), Tam Brown (TB) and Jubrano (Ju) respectively. This suggest that most of the marbles used for this study except Green Pearl (GP) marble may possess potential health threat to the users as observed in Figure 7b, even the Green Pearl (GP) marble in which its values of UAI and ELCR are 1.289 and 0.00131 respectively still need to be assessed before use.

## 4. CONCLUSIONS

Radioisotopes concentration assessments in the imported marbles used in Nigeria for building purposes have been studied using HPGe detector. Radiological risk parameters such as Radium equivalent ( $Ra_{eq}$ ), Dose rate (Dr), Annual Effective Dose Rate (AEDR), Hazard indices ( $H_{in}$  and  $H_{ex}$ ), Gamma index ( $I_\gamma$ ), Alpha index ( $I_\alpha$ ), Utilization Activity Index (UAI) and Excess Lifetime Cancer Risk (ELCR) have been evaluated. The activities concentration of the radioisotopes present in the marbles ranged between 4.30 and 94.47  $\text{Bqkg}^{-1}$ , 42.09 and 104.28  $\text{Bqkg}^{-1}$  and 181.40 and 871.72  $\text{Bqkg}^{-1}$  for  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  respectively. It was observed that there is consistency in the lowest value of radioactivity concentration of  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  obtained for Green Pearl marbles suggesting that this brand of marble may be environmental friendly without pose health challenge.

The radium equivalent of 304.09  $\text{Bqkg}^{-1}$  was found to be Rose Bite marble and the lowest value of 78.46  $\text{Bqkg}^{-1}$  is noted in Green Pearl marble. However, the radium equivalent for each of the marbles is found to be in the range of international reference value of 370  $\text{Bqkg}^{-1}$ . The value of dose rates ranged between 34.94 and 135.68  $\text{nGyh}^{-1}$ . It was observed that the calculated value for dose rate for all the marbles sample used were higher than the international reference value of 80  $\text{nGyh}^{-1}$  except Green Pearl marble. The high values of 1.420, 1.396 and 1.138  $\text{mSv y}^{-1}$  of annual effective dose rate was observed in Rose bite, Black Galaxy and Blue pearl marbles respectively. The values of internal hazard index ranges from 0.223 to 1.077 with Rose Bite and Black Galaxy marbles having the highest value of 1.077

and 1.016 respectively suggesting that Rose Bite and Black Galaxy marbles may not be advisable for use in terms of indoor usage. In terms of external hazard index, it was observed that the values of  $H_{ex}$  in all the samples were less than unity as recommended by [6] suggesting that marbles used for this study could be used for external building construction purposes instead of internal (indoor).

The estimation of gamma activity index value ranged between 0.285 and 1.098. In contrasting the evaluated value of gamma emission index with international reference value when the building materials are applied externally, it was observed that gamma emission index value for Black Galaxy and Rose Bite are within the range whereas in terms of using the material in bulk form marbles such as Blue Pearl, Green Pearl, Jubrano and Tam Brown are within the range of international reference limit. The value of alpha activity index for the samples used ranges from 0.022 to 0.472 with high values of radon concentration noted in Rose Bite and Black Galaxy marbles respectively while lowest is observed in Green Pearl marble. For the utilization of activity index, the highest value of 5.547 is noted in Rose Bite marble while the lowest value of 1.289 is observed in Green Pearl marble. The excess lifetime cancer risk analysis values revealed that the marbles used in this study when compared with the international reference value, it was observed that all the samples have ELCR value that is higher than the mean reference limit value of 0.00029 suggesting that concentration of radio nuclides present in marble should be assessed before use. In conclusion marbles used in this study may mainly be useful for exterior purposes when needed for building project in order to avoid health risk associated with it since 80% of humans spend more time indoor but if there is need be for it to be used especially for indoor purposes marbles such as Rose Bite and Black Galaxy should be excluded. In comparing UAI and ELCR, it was inferred that utmost attention should be paid to the use of Rose Bite, Black Galaxy, Blue Pearl and Tam Brown marbles as a result of high value of excess life cancer risk (ELCR) observed in the samples.

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